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by

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ABSTRACT

Leaves, stems, flowers and seeds of creeping red fescue (Festuca rubra L.), redtop (Agrostis alba L.) and birds foot trefoil (Lotus corniculatus L.) growing on uranium tailings in Elliot Lake were analyzed for traces of radionuclides. The samples were found to contain detectable levels of Ra-226 (1.6 to 10.7 pCi/g), Pb-210 ($\leq$1 to 13.2 pCi/g) and total Pb (<1.0 to 7.0 µg/g). On the other hand other radionuclide levels such as Th-232 (<0.1 pCi/g), Th-230 (<1.0 pCi/g), Th-228 (<0.3 pCi/g), Ra-224 (<0.1 pCi/g) as well as total radionuclide levels such as total Th (<2 µg/g), total U (<0.2 µg/g) were undetectable.

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DOSAGE DES RADIONUCLÉIDES CONTENUS DANS DIVERS TISSUS DE PLANTES POUSSANT SUR DES RÉSIDUS D'URANIUM À ELLIOT LAKE

par

N.K. Davé*, T.P. Lim** et N.R. Cloutier†

RÉSUMÉ

On a fait le dosage des radionucléides présents en traces dans des feuilles, des tiges, des fleurs et des graines de détuques rouges (Fetusca rubra L.), d'agrostis blancs (Agrostis alba L.) et de trèfles connus (Lotus corniculatus L.) poussant sur des résidus d'uranium à Elliot Lake. Les échantillons analysés contenaient des quantités détectables de Ra-226 (1,6 à 10,7 pCi/g), de Pb-210 ($\leq$1 à 13,2 pCi/g) et de Pb total (<1,0 à 7,0 µg/g). Par contre, les radionucléides suivants n'étaient pas présents en concentrations détectables: Th-232 (<0,1 pCi/g), Th-230 (<1,0 pCi/g), Th-228 (<0,3 pCi/g), Ra-224 (<0,1 pCi/g), Th total (<2 µg/g) et U (<0,2 µg/g).

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INTRODUCTION

One of the major terrestrial concerns associated with abandoned uranium tailings is the bioaccumulation of radionuclides by plants growing on tailings and subsequent transfer through the terrestrial food chain (Swanson et al., 1981). Few studies have been conducted on radionuclides uptake by vegetation established on or near uranium tailings. Furthermore information on the distribution of certain radionuclides within various tissues of plants (leaves, stems, flowers, seeds) growing on uranium tailings is practically nonexistent. Such information is greatly needed by radioecologists who are trying to model the migration of radionuclides from tailings to herbivores which may be feeding specifically on grass stems (meadow vole; Peterson, 1966) or grass seeds (jumping mouse; Peterson, 1966). The need for such a study is amplified by the fact that presently most abandoned uranium tailings in Northern Ontario are stabilized by establishing a lush vegetation cover on those waste sites, therefore promoting surrounding indigenous herbivores to feed on tailings.

The present pilot study was designed primarily to determine which radionuclides were sufficiently elevated within various grass and legume tissues, growing on Nordic West Arm tailings, to have the potential of being accumulated at higher trophic levels of the food chain. Secondarily the study was conducted to investigate the distribution of certain radionuclides within the same plant (i.e., leaves, stems, flowers, seeds) since very little information is available on the subject.

METHODS

All samples were collected on the West Arm of the Nordic uranium tailings in Elliot Lake, Ontario. Three to five 1 m^2 quadrants were randomly selected. All of the standing vegetation in each quadrant was cut at ground level, put

in sampling bags and brought back to the laboratory. Sampling dates are shown in Table 1. Substrate core samples (0-47 cm) were also collected from each quadrant.

The vegetation samples were then sorted out according to three different species: creeping red fescue (Festuca rubra L.), redtop (Agrostis alba L.) and birds foot trefoil (Lotus corniculatus L.). The late spring samples were further partitioned into leaves and stems; the summer samples were divided into leaves, stems and flower samples, but part of the original sample was also analyzed whole; only the seeds of the fall samples were kept for analytical purposes (refer to Table 1 for further details). These samples were then washed in a bath of distilled water and thereafter oven dried at 100°C. Following this, composite samples were formed by grouping together similar species of different quadrants. These composite samples were then homogenized and afterwards divided into two parts. One part was wet digested and subsequently analyzed for Pb-210 by counting Bi-210 beta particles after equilibrium and for total Pb by standard atomic absorption technique. The second part of the samples were ashed at 500°C and then digested. These latter digested samples were analyzed for: Th-232, Th-228, Th-230, Ra-226, Ra-224 by alpha spectrometric technique. Total U and total Th were analyzed by neutron activation.

Core samples were divided into five sections according to their depth: 0-5, 5-10, 10-15, 15-30 and 30-47 cm. Composite samples were formed by grouping together similar sections of core samples collected in each quadrant. Composite samples were digested and analyzed as mentioned above for plant tissues.

RESULTS AND DISCUSSION

Table 1 shows that Th-232 and Th-228 were close to the limit of detection in all vegetation samples analyzed. Th-230 was below 1 pCi/g in all

Table 1 - Radionuclide levels in vegetation growing on Nordic uranium tailings based on dry weight (± analytical precision).

Radionuclides	Species	Late Spring (June)		Summer (July)				Fall (Sept)
		Leaves	Stems	Leaves	Stems	Whole	Flowers	Seeds
Th-232 pCi/g	Redtop	0.0±0.0	-	-	-	0.0±0.0	0.0±0 0	-
		-	-	-	-	-	-	-
	Fescue	0.0±0.0	0.0±0.1	0.0±0.0	0.0±0.0	-	0.0±0.0	0.1±0.1
		-	-	-	0.0±0.0	-	0.0±0.0	-
	Trefoil	0.0±0.1	0.0±0.0	0.0±0.0	0.0±0.1	0.0±0.0	0.0±0.0	0.0±0.1
		0.0±0.0	-	-	-	0.0±0.0	-	-
Th-230 pCi/g	Redtop	0.4±0.1	-	-	-	0.0±0.1	0.1±0.1	-
		-	-	-	-	-	-	-
	Fescue	0.3±0.1	0.3±0.1	0.3±0.1	0.1±0.1	-	0.1±0.1	0.2±0.1
		-	-	-	0.1±0.1	-	0.1±0.1	-
	Trefoil	0.2±0.1	0.2±0.1	0.0±0.1	0.0±0.1	0.0±0.1	0.1±0.1	0.7±0.3
		0.1±0.1	-	-	-	0.0±0.1	-	-
Th-228 pCi/g	Redtop	0.0±0.1	-	-	-	0.0±0.1	0.0±0.1	-
		-	-	-	-	-	-	-
	Fescue	0.2±0.1	0.1±0.1	0.1±0.1	0.0±0.0	-	0.0±0.0	0.0±0.1
		-	-	-	0.0±0.0	-	0.1±0.1	-
	Trefoil	0.0±0.1	0.0±0.1	0.2±0.1	0.0±0.1	0.1±0.1	0.0±0.1	0.1±0.2
		0.1±0.1	-	-	-	0.0±0.1	-	-
Total Th u/g	Redtop	<2	-	-	-	<2	-	-
		-	-	-	-	<2	-	-
	Fescue	<2	<2	<2	<2	-	<2	<2
		-	-	<2	<2	-	<2	-
		-	-	-	<2	-	-	-
	Trefoil	<2	<2	-	<2	<2	<2	-
		<2	-	-	-	<2	-	-
PB-210 pCi/g	Redtop	7.5±1.5	-	-	-	8.0±1.5	5.9±1.3	-
		-	-	-	-	-	-	-
	Fescue	3.9±1.3	2.2±1.2	7.8±1.4	3.2±1.2	-	3.8±1.2	8.1±1.3
		-	-	-	2.5±1.1	-	2.4±1.3	-
	Trefoil	7.5±1.5	12.9±1.7	7.6±1.4	13.2±1.7	10.2±1.6	<1±1.0	<1±1.0
		6.4±1.4	-	-	-	12.2±1.7	-	-
Total Pb ug/g	Redtop	2.5	-	-	-	1.0	-	-
		-	-	-	-	1.5	-	-
	Fescue	4.0	1.0	3.5	<1.0	-	2.0	5.0
		-	-	3.0	1.5	-	<1.0	-
		-	-	-	<1.0	-	-	-
	Trefoil	3.0	4.0	-	4.0	5.0	7.0	-
		3.0	-	-	-	1.5	-	-
Ra-226 pCi/g	Redtop	5.3±0.4	-	-	-	10.7±0.8	4.1±0.4	-
		-	-	-	-	-	-	-
	Fescue	4.3±0.4	3.6±0.4	7.2±0.5	4.6±0.3	-	3.9±0.4	3.4±0.3
		-	-	-	4.3±0.3	-	2.5±0.3	-
	Trefoil	2.4±0.3	1.6±0.2	8.7±0.6	2.0±0.3	3.5±0.4	2.1±0.2	6.1±0.7
		2.5±0.3	-	-	-	4.7±0.4	-	-
Ra-224 pCi/g	Redtop	0.1±0.1	-	-	-	0.1±0.1	0.1±0.1	-
		-	-	-	-	-	-	-
	Fescue	0.1±0.1	0.1±0.1	0.1±0.1	0.1±0.1	-	0.1±0.1	0.1±0.1
		-	-	-	0.1±0.1	-	0.1±0.1	-
	Trefoil	0.1±0.1	0.1±0.1	0.1±0.1	0.1±0.1	0.1±0.1	0.1±0.1	0.1±0.1
		0.1±0.1	-	-	-	0.1±0.1	-	-
Total U ug/g	Redtop	<0.2	-	-	-	<0.2	-	-
		-	-	-	-	<0.2	-	-
	Fescue	<0.2	<0.2	<0.2	<0.2	-	0.3	<0.2
		-	-	<0.2	<0.2	-	<0.2	-
		-	-	-	<0.2	-	-	-
	Trefoil	<0.2	<0.2	-	<0.2	<0.2	<0.2	-
		<0.2	-	-	-	<0.2	-	-

samples, while total Th values were all below 2 ug/g (Table 1). These data indicate that Th does not accumulate in specific tissues (i.e., leaves, shoots, flowers, seeds) of grasses and legumes at any measurable levels. Values obtained in this study agree with previous studies, see Table 2.

Table 1 shows that total Pb levels varied between <1.0 to 7.0 ug/g while Pb-210 levels varied between $\leq$1 to 13.2 pCi/g in different tissues of different plant species. The data suggest a rather uniform Pb concentration within all tissues of a plant (i.e., leaves $\gtrsim$ shoots $\gtrsim$ flowers $\gtrsim$ seeds). Furthermore, it can be said that accumulation of Pb-210 in grasses and legumes did not appear to exceed 15 pCi/g while total Pb did not exceed 10 ug/g which agrees with previous studies (Table 2). Ra-224 did not exceed 0.1 pCi/g in all samples (Table 1), and it can therefore be concluded not to accumulate in specific plant tissues to any measurable extent. Ra-226 levels were found to vary from 1.6 to 10.7 pCi/g in grasses and legumes (Table 1). The data indicate that there are no drastic differences in Ra-226 levels between various tissues of grasses and legumes. The range of Ra-226 levels determined in this study agreed with previous work where Ra-226 was found to accumulate in vegetation growing on uranium tailings (Table 2).

Total U levels were below detection limits (0.2 ug/g) in all samples except for one (Table 1) which correlates with previous studies (Table 2).

Note that the root system was not included in this study because of the numerous problems associated with cleaning the samples.

In the substrate it was found that Th isotope levels were substantially lower than Ra and Pb (Th-230 compared to Ra-226 and Pb-210) isotope levels (Table 3). This was believed to be due to the fact that Th is more soluble than Ra and Pb (present in sulphate forms) and hence Th was leached out rather easily. It can therefore be said that the low Th levels measured in tailings,

Table 2 - Radionuclide and total Pb levels in vegetation growing on Nordic uranium tailings and in control samples according to previous studies.

	Moffett et al., 1977*				Cloutier et al., 1983**	
	ug/g (dry wt.)		pCi/g (dry wt.)		ug/g (dry wt.)	pCi/g (dry wt.)
	U	Th	Ra-226	Pb-210	Pb	Ra-226
Elliot Lake Nordic Tailings	.030	.014	5.48	2.32	D.L.-2.0+	9
Control	.010	.017	0.24	0.12	D.L.	0

* Values appearing in the table are mean values of radioisotope concentration of grass (includes values for creeping red fescue, reed canary grass, redtop, climax timothy).

** Values appearing in the table are for mixed grasses and legume samples.

\+ D.L. indicates detection limit.

Table 3 - Radionuclide levels in tailing substrates (Nordic West Arm) collected at different depths from surface. Results are shown for two samples in pCi/g dry weight.

Sample	Depth (cm)	Th-232	Th-230	Th-228	Pb-210	Ra-226	Ra-224
(1)	0-5	1.4±0.6	23.0±2.7	1.9±0.8	170±14	182.7±9.6	3.7±1.4
(2)		1.7±0.5	23.4±2.2	2.8±0.7	133±7	136.9±7	2.1±0.8
(1)	5-10	2.1±0.5	23.5±2.1	5.4±0.9	193±16	180.5±8.4	3.4±1.4
(2)		1.1±0.5	40.0±3.4	4.3±1.0	404±18	182±9.3	2.0±0.9
(1)	10-15	1.5±0.7	17.5±2.4	5.8±1.3	153±13	143.1±8.0	1.3±0.9
(2)		1.8±0.7	76.3±3.0	7.5±1.5	203±10	164.2±8.9	5.0±3.0
(1)	15-30	2.5±0.9	24.5±2.9	6.5±1.4	223±19	202.8±10.1	4.2±1.8
(2)		1.9±0.3	26.1±1.8	5.1±0.6	264±12	196.0±8.5	4.0±1.0
(1)	30-47	1.2±0.7	26.2±3.3	5.3±1.4	226±11	223.4±12.4	6.6±3.2
(2)		3.1±0.9	38.6±3.5	7.7±1.4	226±12	226.9±10.8	9.0±2.0

when compared to Ra and Pb levels, do correlate with the undetectable Th levels found in plants (Table 1). Similarly the relatively higher Ra and Pb levels, when compared to Th levels, measured in tailings do correlate with the higher levels of Ra and Pb measured in plants (Table 1). It is also seen from Table 3 that radionuclide levels are depleted in the upper part of the tailing substrate as a result of leaching as well as building up of a top layer containing decayed organic matter from plants.

CONCLUSIONS

From this pilot study it can first be concluded that grasses and legumes growing on the Nordic West Arm uranium tailings contain measurable levels of Ra-226 (1.6 to 10.7 pCi/g), Pb-210 (<1 to 13.2 pCi/g) and total Pb (<1.0 to 7.0 µg/g). On the other hand other radionuclide levels such as Th-232 (<0.1 pCi/g), Th-230 (<1.0 pCi/g), Th-228 (<0.3 pCi/g), Ra-224 (<0.1 pCi/g) as well as total radionuclide levels such as total Th (<2 µg/g) and total U (<0.2 µg/g) were undetectable. Therefore, it appears that Ra-226, Pb-210 and total Pb should be the primary candidates for future studies on radionuclide accumulation within Elliot Lake uranium tailings ecosystem.

Further, it can be concluded that the levels of Ra-226, Pb-210 and total Pb within leaves, stems, flowers and seeds do not appear to vary to a large extent between various plant tissues.

More data will be collected in the near future in order to complete the statistical analysis.

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